

Amendments to the Specification:

The amendments given below have been numbered for ease of reference to them in the Remarks section of this Reply. The PCT specification does not contain line numbers. The line numbers given below are based on a count of all the lines, including the lines which do not contain words; thus the number of a line at the end of one paragraph, and the number of a line at the end of the next paragraph, differ by a count of 2.

1. On page 1, line 1, please delete the heading ELECTRICAL WIRE INSULATION
2. On page 1, before line 3 (beginning with the words "This invention relates..."), please cancel the amendments requested in the Preliminary Amendment (insertion of the headings Background of the Invention and Field of the Invention), and insert the following new heading

B¹

BACKGROUND OF THE INVENTION

3. Please replace the paragraph beginning on page 1, line 3, with the words "This invention relates..." and ending on page 1, line 10, with the words "... packaging films", with the following amended paragraph.

B²

This invention relates to insulation for electrical wire or cable, (hereinafter "wire") in which a strong bond is achieved at an interface between a layer of a polyolefin-based material and a layer of polyvinylidene fluoride-based material. The invention is especially useful in multi-layer insulation of electrical wires, making it possible to achieve high performance bonding between layers of such materials while retaining an acceptable balance in the complex relationships of other wire performance requirements, which are specialized and different from the criteria for other kinds of articles such as mouldings or packaging films.

4. Please delete the paragraph beginning on page 1, line 12, with the words "The following..." and ending on page 1, line 19, with the words "... vinyl acetate."
5. On page 1, before the paragraph starting on line 21 with the words "Dual wall wire insulation...", please delete the amendment requested in the Preliminary Amendment (insertion of the heading "Description of the Art").

6. Please replace the paragraph beginning on page 1, line 21, with the words "Dual wall wire insulation..." and ending on page 2, line 7, with the words "... bond strength developed." with the following amended paragraph.

B³ ~~Wires~~ Wire and cable with dual wall ~~with chemically resistant dual-wall insulation~~ comprising a polyolefin inner layer (~~core~~) and polyvinylidene fluoride (PVDF) outer layer (~~primary jacket or PJ~~) have been commercially available for over 30 years. In such insulated products, the adhesion between the polyolefin and PVDF layers is negligible, and as a result the products suffer from the certain disadvantages. ~~products,, and are available from several different manufacturers. These products all have negligible adhesion between the inner (polyolefin) and outer (PVDF) layers, which are consequently easily separable. It has been necessary to accept certain disadvantages arising from this lack of bonding, which limits the robustness of the construction. For example, the outer insulation layer can crack and peel of the inner layer is subjected to mechanical stress exposure to certain fluids, contact with sharp objects, or impact. Abrasion and flexural fatigue resistance of the insulation as well as resistance to wrinkling on bending (which can cause difficulties in the ceiling wire or inserting it into profits or connectors) are also detrimental he effected by having two readily separable insulation layers. It has not been for possible to bond layers of two such dissimilar classes of material as polyolefins and PVDF's on a wire and commercially acceptable cost and manufacturing efficiency. Moreover, available bonding techniques could unexpectedly affect the wire performance characteristics. The conventional approach to the bonding of polyolefins and PVDF is to employ a tie layer material (e.g. U.S. Patent 5,589,028), but these tend to be expensive, and when used on wire may compromise other properties, e.g. heat ageing, and add complexity to the manufacturing process in forming extra layer. They may also be of limited effectiveness in terms of bond strength developed.~~

7. Before line 8 on page 2, which begins with the words "It has now been...", please cancel the amendment requested in the Preliminary Amendment (insertion of the heading "Brief Summary of the Invention") and insert the following new heading.

B⁴ SUMMARY OF THE INVENTION

8. Please replace the paragraph beginning on page 2, line 8, with the words "It has now been..." and ending on page 2, line 14, with the words "... performance characteristics." with the following amended paragraph.

B5
It has now been discovered, according to the present invention, that improved insulation can be provided by a first layer comprising a selected carbonyl-containing polymer and an adjacent second layer comprising a selected fluoropolymer. These layers ~~the dissimilar insulation materials of a polyolefin-based core and a polyvinylidene fluoride-based PJ~~ can be bonded together by cross-linking to provide insulation having improved to a significant level of adhesion on an electrical wire or cable; that this bonding tends to reduce or eliminate the aforementioned robustness problems on a wire; and that this bonding can be achieved, contrary to expectation, without unacceptable effects on crack propagation resistance, cost, or on the general balance of wire performance characteristics in one or more areas such as resistance to abrasion, peeling (especially if one of layers is damaged), blistering (especially if heat is applied), delamination, creasing and wrinkling (especially when the insulation is subject to mechanical stress or exposure to solvents).

9. Please delete the paragraph beginning on page 2, line 15, with the words "In the wire..." and ending on page 2, line 18, with the words "... especially ionizing radiation."

B6
10. Please replace the paragraph beginning on page 2, line 21, with the words "The invention accordingly..." and ending on page 3, line 10, with the words "... the uncross-linked layers." with the following amended paragraph.

In a first aspect, this ~~The invention accordingly~~ provides an insulated electrical wire comprising

1) a metallic conductor, and

2) having insulation comprising

B6
(i) at least a first layer which is composed of a polyolefin-based material comprising, of which first polymeric composition consisting of a first polymeric component and optionally a first non-polymeric component, the first polymeric component comprising at least 20%, preferably at least 40%, more preferably at least 60% or preferably at least 80%, by weight, based on the weight of the first polymeric component, (or, in some embodiments, based on the weight of the whole material composition) of a carbonyl-containing polymer (which may be a homopolymer or copolymer or, including terpolymer) having terpolymer, and which preferably has a non-aromatic

B4 backbone), ~~of which polymer the or at least one constituent monomer is the~~ carbonyl-containing polymer comprising repeating units derived from a monomer which (a) can be copolymerized with an olefinic monomer and (b) contains a carboxylic acid ester group, preferably an acrylate or acetate, especially an alkyl acrylate (preferably methyl acrylate, ethyl acrylate, propyl acrylate or butyl acrylate), the units derived from said monomer itself constituting at least 5%, preferably at least 9%, more preferably at least 15%, for example 15 to 28%, by weight of the carbonyl-containing polymer said co-, or ter-polymer when used, and the remainder any other repeating units of the carbonyl-containing polymer said co-, or ter-polymer preferably being derived from an olefinic monomer, preferably ethylene; in contact with;

(ii) at least a second layer which is in direct contact with the first layer at an interface, and which is composed of a second polymeric composition consisting of a second polymeric component and optionally a second non-polymeric component, the second polymeric component comprising of a material containing at least 10%, more preferably at least 50%, particularly or at least 90%, for example substantially 100%, by weight based on the whole material weight of the second composition, of at least one of polyvinylidene fluoride (PVDF), or especially preferably a copolymer based on VDF with a partially or fully fluorinated co-monomer, most preferably a copolymer of VDF and and a vinylidene chloride (VDF) copolymer consisting essentially of

- (a) repeating units derived from vinylidene chloride, and
- (b) repeating units derived from a partially or fully fluorinated co-monomer, preferably hexafluoropropylene (HFP);

the first layer being positioned between the conductor and the second layer. Preferably, the wherein the said layers (i) and (ii), while whilst in contact with each other, have been subjected to conditions which cause cross-linking of polymers at the interface between them reaction, preferably by subjecting the layers to radiation, particularly more preferably ionising radiation. The sufficient to prevent delamination of the two layers during the acetone emotion test described below, or to increase cross-linking is preferably such that at least one of the following conditions is fulfilled

- (a) the peel bond strength between the said layers, measured by ASTM 81876- 95, is to at least 5N, preferably more than 10N according to the ASTM 1876-95 method described below preferably increasing the,

B6
could

(b) when a sample of the insulated electrical wire 60 mm long is immersed in a bath of acetone 4.2 mm deep at 23 °C for 1 hour, there is no delamination of the two layers, and

(c) the peel bond strength between the layers after the crosslinking, measured by ASTM B1876-95, is at least 50%, more preferably by at least 100%, especially by at least 500% or 1000%, compared to that between the uncross-linked layers greater than the peel bond strength between the layers before the crosslinking, measured by ASTM B1876-95.

11. Please add the following new paragraph after the amended paragraph set out in item 10 above.

B7

Throughout this specification, including the claims, the terms "a", "an" and "the" before an item mean that there can be a single such item or two or more such items, unless the context makes this impossible (for example, in the first aspect of the invention, the first polymeric component can comprise a single carbonyl-containing polymer as defined or two or more such polymers; and the second polymeric component can contain a single fluoropolymer or a mixture of two or more fluoropolymers); and the term "consisting essentially of" certain ingredients means that those ingredients are necessarily present and that other ingredients may be present providing that their presence does not substantially change the properties of the insulation.

12. Please replace the paragraph beginning on page 3, line 12, with the words "According to another aspect of the invention..." and ending on page 4, line 4, with the words "... the uncrosslinked layers." with the following amended paragraph.

A second According to another aspect of the invention provides a method of making an insulated wire or cable, the method comprising the steps of

B8

(A) providing an electrical conductor surrounded by

(i) a first layer which is composed of a first polymeric composition as defined in the first aspect of the invention; and

(ii) a second layer which is composed of a second polymeric composition as defined in the first aspect of the invention;

the first and second layers being in direct contact with each other at an interface, and the first layer being positioned between the conductor and the second one; and

(B) exposing the layers while in contact with each other to ionising radiation which causes cross-linking of polymers at the interface.

we provide an electrical wire having insulation comprising:

- B8
amended
- (i) at least a first layer of a polyolefin-based formulation, of which at least 20%, preferably at least 40%, more preferably at least 60% or very preferably at least 80% of the weight of the polymeric portion of the said formulation consists of a carbonyl-containing polymer (homopolymer or copolymer or terpolymer), of which polymer the or at least one constituent monomer is a carboxylic acid ester, preferably an acrylate or acetate, especially an alkyl acrylate (preferably methyl acrylate, ethyl acrylate, propyl acrylate or butyl acrylate), the said monomer itself constituting at least 5%, preferably at least 9%, more preferably at least 15% by weight of the said co-, or ter- polymer when used, and the remainder or the majority of the remainder of the said co-, or ter- polymer preferably being derived from olefinic monomer, preferably ethylene; in contact with
- (ii) at least a second layer of another material formulation, containing at least 10%, more preferably at least 50%, very preferably at least 90%, especially 100%, by weight of the second layer, of polyvinylidene fluoride (PVDF), or especially preferably of a copolymer based on VDF with a partially or fully fluorinated co-monomer, most preferably a copolymer of VDF and hexafluoropropylene (HFP);

wherein the said layers (i) and (ii) whilst in contact with each other have been subjected to cross-linking reaction, preferably by radiation, more preferably ionising radiation, sufficient to prevent delamination of the two layers during the acetone immersion test described below, or to increase the peel bond strength between the said layers to at least 5N according to the ASTM B1876-95 method described below preferably increasing the bond strength by at least 50%, more preferably by at least 100%, especially by at least 500% or 1000%, compared to that between the uncrosslinked layers.

13. After the amended paragraph set out in item 12 above, please insert the following new heading.

B9
DETAILED DESCRIPTION OF THE INVENTION

14. After the new heading set out in item 13 above, please insert the following new paragraph.

B10
When the first polymeric component does not consist solely of the carbonyl-containing polymer, any other polymer present in the first polymeric component is preferably a polyolefin, particularly high-density polyethylene.

15. Please delete the paragraph beginning on page 4, line 6, with the words "Preferably, the respective layers..." and ending on page 4, line 10, with the words "... cross-linking reaction."

16. Please replace the paragraph beginning on page 4, line 12, with the words "The polyolefin-based layer ..." and ending on page 4, line 15, with the words "... properties to the polymer." with the following amended paragraph.

B11
Each of the layers (i) and (ii) optionally contains, in ~~The polyolefin-based layer (i)~~ in addition to the polymeric component of the composition, a non-polymeric component comprising portion of the formulation, for which the requirements are stipulated above, may ~~contain whatever else is required in the way of~~ additives such as anti-oxidants, pigments, fillers, flame retardants, etc, to enhance as known per se, to give the required mechanical, thermal, electrical etc. properties of the insulation to the polymer.

17. Please delete the paragraph beginning on page 4, line 17, with the words "The polyvinylidene fluoride-based layer..." and ending on page 4, line 18, with the words "... in addition to bonding."

18. Please delete the paragraph beginning on page 4, line 20, with the words "Advantages of achieving ..." and ending on page 4, line 26, with the words "... the inner layer."

19. Please replace the paragraph beginning on page 5, line 1, with the words "The bond strength described..." and ending on page 5, line 30, with the words "... under a microscope." with the following amended paragraph.

B12
The bond strength described in this application can be measured in terms of peel strength ~~between bonded strips of the two materials in question. A standard method which can be used for such a test is ASTM 1876-95. By this definition, a significant bond could be one for which the peel force exceeds 5N, and a strong bond one of peel force greater than 10N.~~ A convenient method for gauging the bond strength between the layers said layers, (i) and (ii), when they have been fabricated onto a wire, is to place a sample wire, of total length 60mm, into acetone (e.g. Fisher Scientific UK, AR certified grade acetone), to a depth of acetone equivalent to 70% of the length of sample wire, at 23 (+/- 3)°C, for a period of 1 hour. Wires with negligible bonding of the insulation layers experience an extension of the outer layer PVDF-PJ, along the axis of the wire, that is independent of any extension of the inner layer polyolefin core, and/or wrinkling of the outer layer PJ such that it delaminates

B12
cancel

from the inner layer core in places. When it occurs, the above-mentioned extension of the outer layer PJ typically results in a PJ "tube" extending for 1mm or more beyond the cut end of the inner layer core in the sample wire, following the above test. Wires with significantly bonded insulation layers experience an extension of both layers the core and PJ, together, without separation, beyond the cut edge of the conductor, along the axis of the wire and/or wrinkling of the two core and PJ layers together, without delamination. Any such wrinkling of the two layers core and PJ together can be distinguished from wrinkling only of the outer layer PJ only by examining a cross-section of the wrinkles under a microscope.

20. Please replace the paragraph beginning on page 5, line 20, with the words "Methods of fabricating the wire..." and ending on page 5, line 30, with the words "... pre-formed underlying layer." with the following amended paragraph.

B13

In the method of the invention, step (A) can make use of any ~~Methods of fabricating the wire may include any~~ process which causes intimate contact between the above-mentioned layers (i) and (ii). Examples include coating the second polymeric composition of one material onto a pre-formed layer of the first polymeric composition ~~ether, and~~ dual or multi-walled extrusion to form insulation layers respectively containing one or other of the polymeric compositions, aforementioned two classes of material. ~~The olefin-based material (i) is preferably the inner layer and the PVDF-based layer is preferably the outer layer on the wire.~~ The layers made from the two different compositions can materials could be coextruded, tandem extruded, multipass extruded, or coated by other means. Known wire insulation processes such as tube draw-down extrusion may be used, to form one or more of the layers, but pressure extrusion as known per se is preferred for optimum adhesion of the second and any subsequent insulation layers to be applied to a pre-formed underlying layer. In some embodiments of the invention, the first layer is in direct contact with the conductor. In some embodiments of the invention, the defined first and second layers are the sole insulation around the conductor. In other embodiments, the first and second layers are part of multilayer insulation including one or more other layers.

21. Please replace the paragraph beginning on page 6, line 1, with the words "... The insulation on the wire" and ending on page 6, line 12, with the words "... into the material." with the following amended paragraph.

B14

In step (B) of the method of the invention, the ~~The~~ insulation on the wire is exposed to conditions which cause a cross-linking reaction. The cross-linking, which may involve chemical reagents such as peroxides, but preferably is effected by radiation, especially

B14
cond

radiation from a source of ionising radiation capable ~~source of ionising radiation capable~~ of causing the formation of free radicals and thus, cross-links, in the polymers, some of which should preferably be formed in the region of the interface between the two materials compositions. Penetration of the radiation into the material insulation at least as far as the interface is therefore desirable, although not necessarily essential if ion or radical mobility, for example, enables molecular reactions to continue at or near the interface after the radiation process. The radiation source could, for example, be a radio-isotope, or an X-ray source, or possibly a non-ionising radical-generating source, for example a UV source, but is preferably an electron beam, more preferably one providing a beam dose greater than 2 Mrads, preferably at least 5 Mrads, more preferably at least 10 Mrads, very preferably at least 15 Mrads, into the material.

22. Please replace the paragraph beginning on page 6, line 14, with the words "It has been found" and ending on page 6, line 19, with the words "... PVDF-based material." with the following replacement paragraph.

B15

It has been found that, when the cross-linking is effected by ionizing radiation, enhancements to the interfacial bond strength may be obtained by including ~~using certain additives.~~ Additives preferably include a cross-linking promoter ("pro-rad") in the first and/or second polymeric composition. ~~polyolefin-based material and/or in the PVDF-based material.~~ Known pro-rads ~~cross-linking materials~~ may be used, preferably methacrylate/acrylate based pro-rads, e.g. ones, and, very preferably, those of the type trimethylolpropanetrimethacrylate (TMPTM) ~~in the polyolefin material and/or in the PVDF-based material.~~

23. After page 7, line 2 (which ends with the words "... pressure as above"), please insert the following new paragraph.

B16

In the Experimental Results shown below, the first composition (comprising the carbonyl-containing polymer) is referred to as the polyolefin-based material and as Material 1, and the second composition is referred to as the PVDF-based material and as Material 2; and the following abbreviations are used (in addition to those already given). EVA is ethylene/vinyl acetate copolymer. VA is vinyl acetate. EEA is ethylene/ethyl acrylate copolymer. EA is ethyl acrylate. EMA is ethylene/methyl acrylate copolymer. MA is methyl acrylate. HDPE is high-density polyethylene. PVDF is polyvinylidene fluoride.